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# On the decay of $Bi^{204}$

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With 10 figures in the text

## ABSTRACT

Energy levels in $Pb^{204}$ have been studied in the electron capture decay of 12 h $Bi^{204}$. 45 transitions and, in addition, about 20 weak, unassigned conversion lines were found in this very complex decay. Energies and intensities of conversion lines were measured in permanent magnet and double-focusing spectrometers. Relative gamma intensities were obtained by use of a scintillation spectrometer. Electron-gamma and gamma-gamma coincidence measurements were performed with instruments including a 100-channel pulse-height analyzer. A tentative decay scheme is discussed.

## 1. Introduction

The work described here lies within the general frame-work of investigations concerning the nuclei in the vicinity of the double-magic $Pb^{208}$ and was begun in 1955. The shell theory of the nucleus could be used to describe the general characteristics of the level schemes based on experimental measurements for the nuclei $Pb^{207}$ [1], $Pb^{206}$ [2] and $Pb^{205}$ [3] (differing by 1, 2 and 3 neutrons, respectively, from the double-closed shell of $Pb^{208}$). Theoretical calculations by True [4] and more recently by Blomqvist [5] on the energy levels in $Pb^{204}$ added further interest to the experimental investigation of this nucleus. A comparison between theoretical and experimental results is, however, difficult in this case because of the complexity of the level scheme due to the large number of neutron holes (four). Nevertheless, the level scheme of the 67.5 min isomer $Pb^{204m}$, studied by Herrlander *et al.* [6], could be understood in terms of the shell model. The possibility of obtaining a further check of theory from an experimental investigation of the levels in $Pb^{204}$ may therefore not be completely excluded.

Some preliminary data from this investigation were reported in 1956 [7] and 1957 [8]. Although the work is not yet completed, the experimental results obtained so far will be given here in more detail. 45 transitions, and in addition about 20 weak conversion lines due to unassigned transitions, were found.

During the course of the present investigation, Wertheim and Pound [9] have published results on the decay of Bi$^{204}$. Eight gamma rays were found, two of which were not observed in our measurements. They also performed gamma-gamma coincidence measurements. Furthermore, Fritsch [10] in his doctor's thesis includes results on the same decay. 35 transitions in the "first confidence group" and further 32 transitions in the second and third groups are reported. His results, obtained mainly from measurements in permanent magnet beta-spectrometers for photographic recording, are mostly in satisfactory agreement with our results from the corresponding measurements.

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## 2. Experimental procedures

The activities of 12 h Bi$^{204}$ were generally produced in proton bombardments on radiogenic lead (containing 88 % Pb$^{206}$) in the synchrocyclotron of the Gustaf Werner Institute for Nuclear Chemistry. Because Bi$^{203}$, which has about the same half-life, is also produced in these bombardments, different bombarding energies (30–60 MeV) could be used to distinguish between the two isotopes (see also ref. 11). However, in order to make more definite isotopic assignments of gamma rays, Bi$^{204}$ was produced by bombarding spectroscopically pure Tl with 38 MeV alpha-particles in the 225 cm cyclotron of the Nobel Institute. No observable amount of Bi$^{203}$ was produced in these bombardments (cf. Fig. 1 and Section 3).

After chemical separation, Bi sources were prepared for investigations in different spectrometers (see 2.1). In some cases, mainly for measurements of coincidences and scintillation spectra, electromagnetically separated Bi$^{204}$ samples were used.

Energy determinations were made in a permanent magnet beta-spectrometer for photographic recording [12] and in a double-focusing beta-spectrometer [13]. Relative conversion line intensities were in most cases obtained from measurements in the double-focusing beta-spectrometer.

A single-channel scintillation spectrometer with automatic recording [14] was used in order to estimate relative gamma intensities and to search for high-energy gamma rays.

Electron-gamma coincidence measurements were performed with an apparatus including an intermediate-image beta-spectrometer [15], a 100-channel pulse-height analyzer [16] and a coincidence unit [17].

A search for positrons was made in an intermediate-image beta-spectrometer [15] and a double-focusing spectrometer [13].

### 2.1 *Source preparation*

(*a*) The separation of the active bismuth from the target material (in the case of proton-bombardments on lead) and the preparation of sources have been described in previous works [7, 3, 18].

(*b*) In those cases when the bismuth was produced from alpha-bombardments on thallium, the activity was separated in the following way: The thallium was dissolved in a minimum amount of $HNO_3$, and carriers, 3 mg $Fe(NO_3)_3$ and 5 $\mu g$ $Bi(NO_3)_3$, were added. $Tl^{3+}$ was reduced by an excess of $H_2O_2$ to $Tl^+$, and the hydroxides of Fe and Bi were precipitated in the diluted solution by ammonia. The centrifuged precipitate was dissolved in a drop of conc. $HNO_3$ and a second reduction–precipitation cycle was made as above. The precipitate was centrifuged, washed three times with distilled water and redissolved with 6 *N* HCl. The Fe carrier was then separated from the active bismuth by solvent extraction using ether saturated with 6 $N$ HCl. The aqueous phase containing the bismuth was evaporated to a convenient volume. The source preparation is described elsewhere [7, 3, 18].

(*c*) The isotope-separated samples were prepared from the bismuth activities obtained from proton bombardments on lead. The chemical separation was accomplished mainly as in *a*, above. In this case, however, the activity was washed off the ion-exchange column with 1 $N$ $HNO_3$ (instead of $H_2SO_4$) and boiled with HCl until no traces of nitrate remained. After addition of carrier, $BiCl_3$, the electromagnetic isotope separation [19] was performed on the dried substance, the bismuth being collected on a thin foil of lead. After a radiographic determination of the accurate positions of the collected isotopes, a stripe containing $Bi^{204}$ was cut off and the chemical separation of the bismuth activity from the lead was undertaken as in *a*, above.

## 3. Internal conversion line spectra

Fig. 1 shows some of the photographic films which were obtained with the permanent magnet beta-spectrometer. The activities used for these exposures were produced in alpha- and proton-bombardments. The conversion line spectrum was measured also in the double-focusing beta-spectrometer with resolutions varying between 0.15 and 0.5 %. Parts of these measurements are represented in Figs. 2 and 3.

Table 1 includes the transitions in $Bi^{204}$ which were assigned from the measured conversion lines. Each of these lines has been shown to belong to the decay of $Bi^{204}$ from its 12 h half-life and from the fact that it was observed in activities produced in the 38 MeV alpha-bombardment on Tl. As was pointed out above, no observable amount of $Bi^{203}$ was produced in this bombardment. This is proved by the fact that the strong conversion lines of the 825.2 keV transition in $Bi^{203}$ were completely lacking on the photographic film, as can be seen in Fig. 1. In two exceptional cases, the 1202.9 and 1210.8 keV transitions, the isotopic assignment was made by the method of using different bombarding energies. For the stronger transitions the isotopic assignment could be checked from conversion measurements with electromagnetically isotope-separated samples (cf. Fig. 6).

The energy values in Table 1 should be accurate to better than 1:1000. The well-known transitions of $374.7 \pm 0.4$ keV and $911.7 \pm 0.3$ keV [6] were used for calibration. Table 1 includes also relative conversion line intensities (in most cases considered to be accurate to about 15 %) and conversion ratios. For some of the weakest lines, marked $^a$, these data were obtained from visual comparisons on the photographic film and should be considered as rough estimates.

In Table 3 the weak conversion lines belonging to $Bi^{204}$ are given for which the assignment is uncertain. No corresponding $K$ and $L$ lines were found for these transitions.

The transitions given in Table 1 were found also by Fritsch [10], except for a few of them, namely the 90.9, 92.2, 368.0, 473.4, 692.0, 708.8 and 983.6 keV transitions. The corresponding conversion lines were either interpreted in a different way, or assigned to another isotope or not observed at all. In the case of the 983.6 keV transition, which is also reported by Wertheim and Pound [9], our $L$ 983.6 is interpreted by Fritsch as a $K$ line. $K$ 983.6 would almost coincide with the strong $L$ line of the 911.7 keV transition, and may therefore be undetected in the measurements, even at the high resolution used. From measurements in the double-focusing spectrometer at 0.16 % resolution the ratio $K$ 911.7/$L$ 911.7 was found to be $1.60 \pm 0.10$

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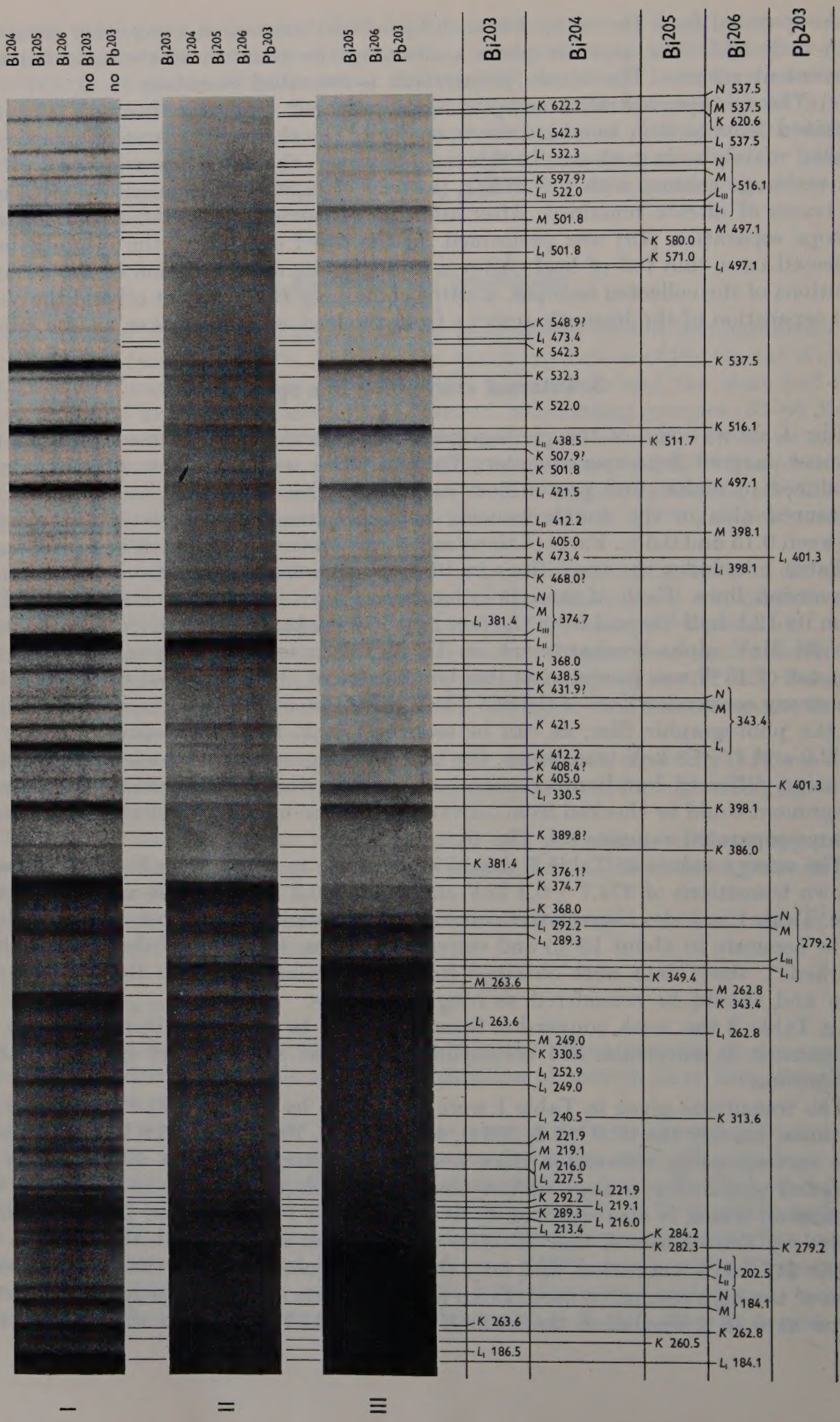

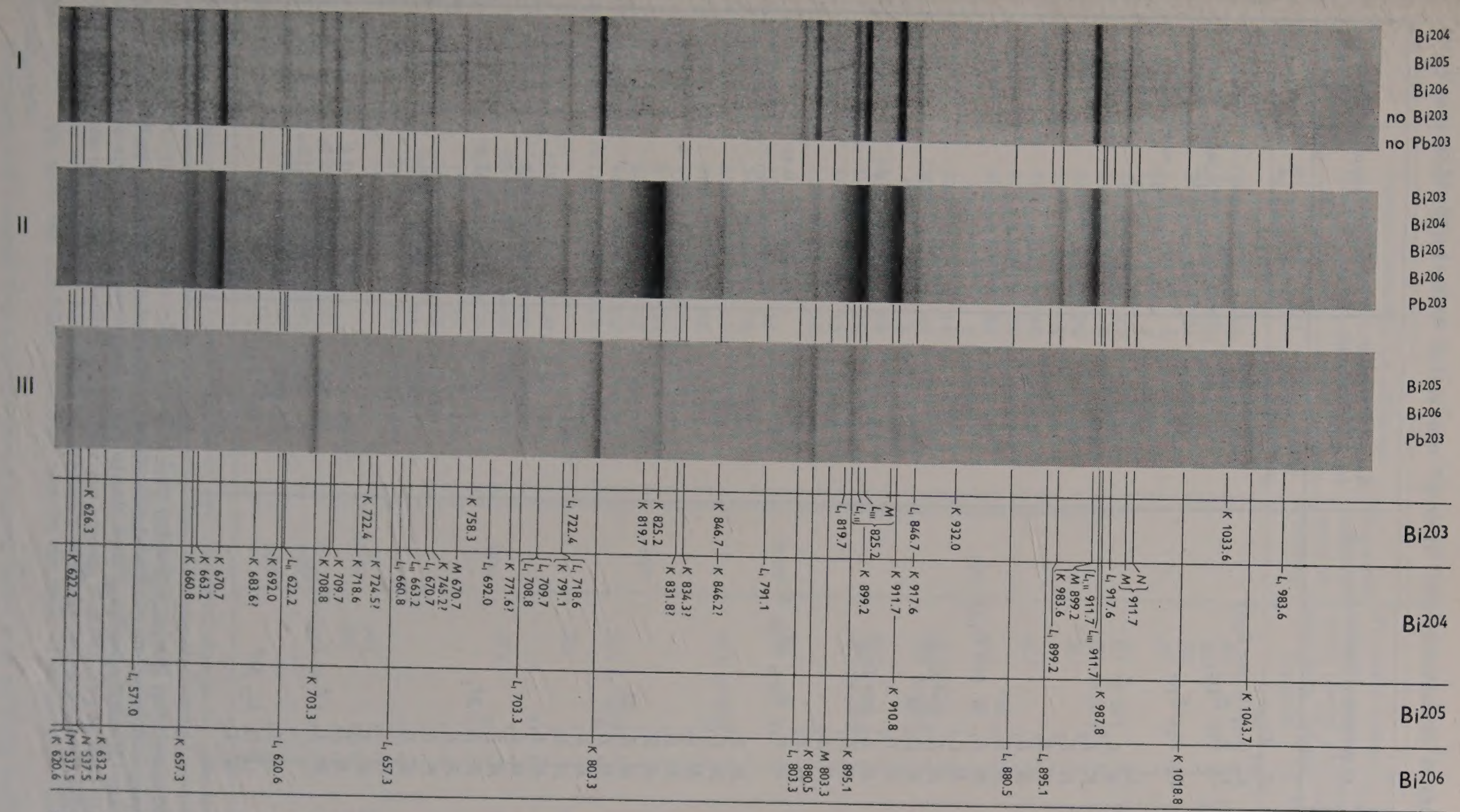

Fig. 1. Energy region 150–1000 keV of conversion line spectra, recorded from about 12 hours' exposures in a permanent magnet spectrometer. The samples were produced in (I) an alpha-bombardment on Tl; (II and III) a proton-bombardment on Pb. The isotopic assignment of the conversion lines belonging to the decays of $Bi^{203}$ and $Bi^{204}$ (both with about 12 hours' half-life) is made from a comparison between the films: *Film No. I* includes lines from $Bi^{204}$ (and the longlived $Bi^{205}$ and $Bi^{206}$) but no lines belonging to the decay of $Bi^{203}$ (*K* 825.2, the strongest conversion line in $Bi^{203}$, is completely lacking, as can be seen). *Film No. II* contains lines from $Bi^{203}$ and $Bi^{204}$ (and $Pb^{203}$, $Bi^{205}$ and $Bi^{206}$). *Film No. III* was exposed 2 days later than film No. II, and shows therefore lines mainly from 52 h $Pb^{203}$, 14.5 d $Bi^{205}$ and 6.4 d $Bi^{206}$.

*Table 1.* Gamma transitions and internal conversion lines in the decay of $Bi^{204}$.

| Transition energy (keV) | Conversion lines observed | $K$-line intensity ($K$ 374.7 = 100) | Conversion ratios |
|---|---|---|---|
| 78.54 | $L_{II}$, $L_{III}$ | $\approx 150\ (\Sigma L)$ | $L_{II}/L_{III} \approx 1.3^{a}$ |
| 80.2 | $L_{I}$, ($L_{II}$?), (no $L_{III}$) | $\approx 60\ (\Sigma L)$ | |
| 90.9 | $L_{I}$, $L_{II}$, $L_{III}$ | $35\ (\Sigma L)$ | $L_{I}/L_{II} \approx 1.3^{a}$;<br>$(L_{I}+L_{II})/L_{III} \approx 4^{a}$ |
| 92.2 | $L_{I}$, $L_{II}$, $L_{III}$ | $10\ (\Sigma L)$ | $L_{I}/L_{II} \approx 2^{a}$;<br>$(L_{I}+L_{II})/L_{III} \approx 4^{a}$ |
| 100.2 | $L_{I}$ | 9 ($L_{I}$) | |
| 140.9 | $K$, $L_{I}$ | 8 ($L_{I}$) | $K/L \approx 5^{a}$ |
| 170.2 | $K$, $L_{I}$ | 10 | $K/L = 6 \pm 2$ |
| 175.9 | $K$, $L_{I}$ | 50 | $K/L \approx 5^{a}$ |
| 213.4 | $K$, $L_{I}$ | 7 | |
| 216.0 | $K$, $L_{I}$ | 41 | $K/L = 6 \pm 2$ |
| 219.1 | $K$, $L_{I}$, $M$ | 13 | $K/L = 4 \pm 2$ |
| 221.9 | $K$, $L_{I}$, $M$, (no $L_{III}$) | 24 | $K/L = 6 \pm 2$ |
| 227.5 | $K$, $L_{I}$ | 1.8 | |
| 240.5 | $K$, $L_{I}$, (no $L_{III}$) | 9 | $K/L = 6 \pm 2$ |
| 249.0 | $K$, $L_{I}$, $M$ | 47 | $K/L = 5 \pm 1.5$ |
| 252.9 | $K$, $L_{I}$ | 3.5 | |
| 289.3 | $K$, $L_{I}$, (no $L_{III}$) | 38 | $K/L = 5.8 \pm 1$ |
| 292.2 | $K$, $L_{I}$ | 5 | $K/L \approx 5^{a}$ |
| 330.5 | $K$, $L_{II}$ | 3.4 | $K/L \approx 5^{a}$ |
| 368.0 | $K$, $L_{I}$ | 2 | $K/L \approx 2^{a}$ |
| 374.7 | $K$, $L_{II}$, $L_{III}$, $M_{II}$, $N$ | 100 | $K/L = 2.4 \pm 0.4$;<br>$(L_{I}+L_{II})/L_{III} = 4.1 \pm 0.4$ |
| 405.0 | $K$, $L_{I}$ | 2.3 | $K/L \approx 3^{a}$ |
| 412.2 | $K$, $L_{II}$ | 2.8 | $K/L \approx 4^{a}$ |
| 421.5 | $K$, $L_{I}$ | 7.1 | $K/L = 6 \pm 2$ |
| 438.5 | $K$, $L_{I}$ | 4.1 | $K/L \approx 6^{a}$ |
| 473.4 | $K$, $L_{I}$ | $\approx 1^{a}$ | |
| 501.8 | $K$, $L_{I}$, $M$ | 3.2 | $K/L \approx 4^{a}$ |
| 522.0 | $K$, $L_{II}$ | 2.5 | |
| 532.3 | $K$, $L_{I}$ | 5.9 | $K/L = 6 \pm 2$ |
| 542.3 | $K$, $L_{I}$ | $\approx 1$ | |
| $622.2^{b}$ | $K$, $L_{II}$ | $< 1$ | |
| 660.8 | $K$, $L_{I}$ | 5.0 | $K/L = 5 \pm 1.5$ |
| 663.2 | $K$, $L_{II}$ | 1.8 | |
| 670.7 | $K$, $L_{I}$, $M_{I}$ | 19 | $K/L = 5.8 \pm 0.6$ |
| 692.0 | $K$, $L_{I}$ | $\approx 1$ | |
| 708.8 | $K$, $L_{I}$ | 3.0 | $\Big\}\ \Sigma K/\Sigma L \approx 3$ |
| 709.7 | $K$, $L_{I}$ | 1.5 | |
| 718.6 | $K$, $L_{I}$ | 2.1 | |
| 791.1 | $K$, $L_{I}$ | 2.3 | |
| 899.2 | $K$, $L_{I}$ | 23 | $K/L = 4.4 \pm 0.4$ |
| 911.7 | $K$, $L_{I}+L_{II}$, $L_{III}$, $M_{II}$, $N$ | 36 | $K/L = 1.6 \pm 0.2$;<br>$(L_{I}+L_{II})/L_{III} = 14 \pm 3$ |
| 917.6 | $K$, $L_{I}$ | 2.9 | $K/L \approx 5^{a}$ |
| 983.6 | ($K$?), $L_{I}$, $M^{c}$ | 0.7 ($L_{I}$) | |
| 1202.9 | $K$, $L_{I}$ | 0.6 | $K/L = 5 \pm 1$ |
| 1210.8 | $K$ | 0.1 | |

[a] Visual estimate from the photographic film.

[b] The conversion lines on the limit of observability. In the decay of 67.5-min $Pb^{204m}$, however, the $K$-, $L_{II}$- and $M$-conversion lines were clearly observable.

[c] The photo peak of a ca. 980 keV gamma ray visible in scintillation spectrum and in electron-gamma coincidence spectra (see e.g. Figs. 7 and 8).

*Table 2.* Multipolarities and intensities of transitions in the decay of $Bi^{204}$.

| Transition energy (keV) | Gamma intensity for different multipolarities, calculated from *K*- (or *L*-) line intensity | | | | | Measured relative gamma intensity[a] | Probable multipolarity assignment (based on data from Tables 1 and 2) | Total transition intensity adopted from *K*- (or *L*-) line intensity |
|---|---|---|---|---|---|---|---|---|
| | *E*1 | *E*2 | *M*1 | *M*2 | *E*5 | | | |
| 78.54 | ≈1200 | ≈10 | ≈60 | ≈3 | | | *E*2 | ≈200 |
| 80.2 | ≈500 | ≈5 | ≈25 | ≈1 | | | *M*1 | ≈450 |
| 90.9 | 390 | 4 | 21 | 1 | | | *E*1 | 600 |
| 92.2 | 115 | 1 | 6 | 0.3 | | | *E*1 | 180 |
| 100.2 | 130 | 2 | 7 | 0.4 | | | | |
| 140.9 | 290 | 9 | 16 | 2 | | 400 | *E*1 or *M*1 | 340 or 70 |
| 170.2 | 110 | 42 | 6 | 1 | | | *E*1 or *M*1 | 120 or 18 |
| 175.9 | 580 | 230 | 33 | 7 | | | *M*1 | 95 |
| 213.4 | 130 | 50 | 8 | 2 | | | | |
| 216.0 | 790 | 300 | 48 | 12 | | | *M*1 | 100 |
| 219.1 | 250 | 96 | 15 | 4 | | | | |
| 221.9 | 490 | 190 | 30 | 7 | | | *M*1 | 60 |
| 227.5 | 39 | 15 | 2 | 1 | | | | |
| 240.5 | 230 | 84 | 14 | 4 | | | *M*1 (*E*1) | 25 |
| 249.0 | 1300 | 470 | 80 | 21 | | | *M*1 or *M*2 | 140 or 80 |
| 252.9 | 97 | 36 | 6 | 2 | | | | |
| 289.3 | 1430 | 540 | 99 | 28 | | | *M*1 | 140 |
| 292.2 | 190 | 72 | 13 | 4 | | | *M*1 or *M*2 | 19 or 10 |
| 330.5 | 180 | 65 | 13 | 4 | | 2600 | | |
| 368.0 | 130 | 49 | 10 | 3 | | | | |
| 374.7 | | 2600 | | | | | *E*2 | 2700 |
| 405.0 | 190 | 70 | 15 | 5 | | | | |
| 412.2 | 240 | 89 | 19 | 6 | | | | |
| 421.5 | 630 | 240 | 51 | 17 | | | *M*1 or *M*2 | 60 or 26 |
| 438.5 | 390 | 150 | 33 | 11 | | 200 | | |
| 473.4 | ≈110 | ≈40 | ≈10 | ≈3 | | | | |
| 501.8 | 410 | 160 | 36 | 13 | | | | |
| 522.0 | 350 | 130 | 32 | 12 | | | | |
| 532.3 | 840 | 330 | 79 | 29 | | | *M*1 or *M*2 | 90 or 40 |
| 542.3 | ≈150 | ≈60 | ≈15 | ≈5 | | | | |
| 622.2 | | | | | 2[b] | 800 | *E*5 | 2[b] |
| 660.8 | 1100 | 420 | 120 | 45 | | | | |
| 663.2 | 400 | 150 | 43 | 16 | | | | |
| 670.7 | 4300 | 1700 | 460 | 180 | | | *M*1 | 490 |
| 692.0 | ≈240 | ≈90 | ≈25 | ≈10 | | | | |
| 708.8 | 760 | 290 | 84 | 33 | | | | |
| 709.7 | 380 | 150 | 42 | 17 | | | | |
| 718.6 | 550 | 210 | 61 | 24 | | | | |
| 791.1 | 710 | 270 | 84 | 34 | | 5000 | | |
| 899.2 | | 3500 | | | | | *E*2 | 3500 |
| 911.7 | | | | | 660 | | *E*5 | 720 |
| 917.6 | 1200 | 460 | 160 | 66 | | | | |
| 983.6 | 2100 | 630 | 270 | 100 | | 400[c] | | |
| 1202.9 | 390 | 160 | 65 | 28 | | 200 | | |
| 1210.8 | 66 | 26 | 11 | 5 | | | | |

[a] The values were obtained from scintillation spectrum and should be considered as rough estimates. Because of the weakness of the isotope separated sample this had to be placed near the crystal. For this reason the intensity values in column 3 may have been somewhat influenced by sum-up effects. The values have been arbitrarily fitted (by multiplication with a constant factor) to give approximate agreement between the values in columns 2 and 3 for the 374.7 keV transition.

[b] The value was calculated by use of the intensity relations between the transitions in 67.5 min $Pb^{204m}$ [6], since the 622.2 keV transition was on the limit of observability in the present investigation.

[c] This value was obtained from a scintillation spectrum taken with an X-ray absorber inserted between the source and the crystal.

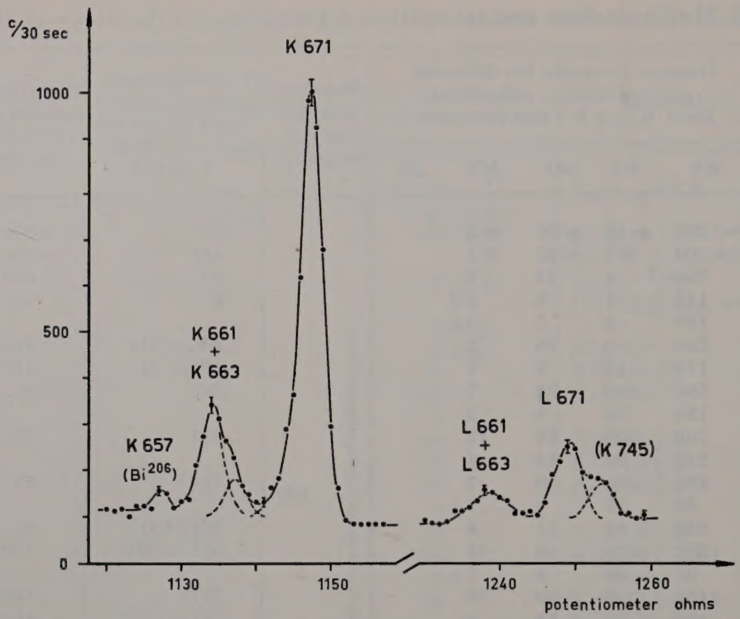

Fig. 2. Part of the conversion-electron spectrum of Bi$^{204}$ measured in the double-focusing spectrometer at 0.16 % resolution. The spectrum is corrected for decay.

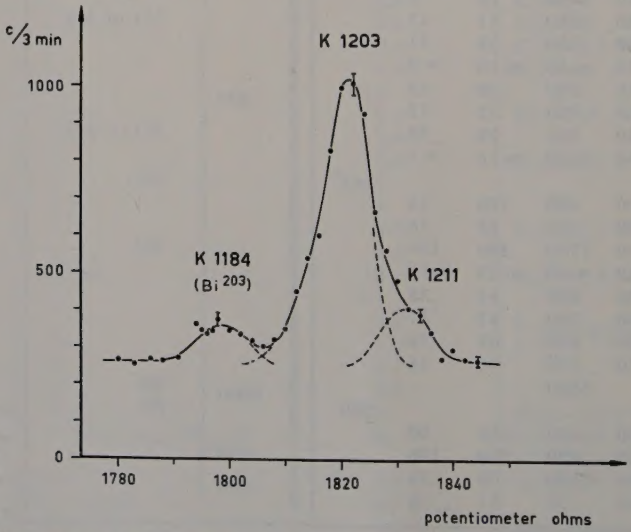

Fig. 3. *K*-conversion lines of the 1202.9 and 1210.8 keV transitions in Bi$^{204}$ measured at 0.5 % resolution in the double-focusing spectrometer. Corrected for decay.

*Table 3.* Unassigned conversion lines in the decay of $Bi^{204}$.

| Electron energy (keV) | Approximate intensity ($K$ 374.7 = 100) | Possible assignment |
|---|---|---|
| 121.0 | 2 | $K$ 209.0 |
| 123.2 | 8 | $K$ 211.2 |
| 136.8 | 2.3 | $K$ 224.8 |
| 288.1 | 4 | $K$ 376.1 or $L_{\mathrm{I}}$ 304,0[a] |
| 301.8 | 1 | $K$ 389.8 |
| 320.4 | 1 | $K$ 408.4 |
| 343.9 | 1 | $K$ 431.9 |
| 380.0 | 2 | $K$ 468.0 |
| 419.9 | $<1$ | $K$ 507.9 |
| 460.9 | 1.5 | $K$ 548.9 |
| 509.9 | 1 | $K$ 597.9 |
| 595.6 | 1 | $K$ 683.6 |
| 636.5 | 1 | $K$ 724.5 |
| 657.2 | 1.6 | $K$ 745.2 |
| 683.6 | 1 | $K$ 771.6 |
| 743.8 | 1.5 | $K$ 831.8 |
| 746.3 | 1.5 | $K$ 834.3 |
| 758.2 | 1.5 | $K$ 846.2 |

[a] $K$304.0 may overlap $M$219.1.

(the overlapping $M$ 899.2 being subtracted), whereas the same ratio in the decay of $Pb^{204m}$ was found to be $1.71 \pm 0.08$ [6]. The theoretical value [20] of a 911.7 keV $E5$ transition is 1.77. This result does not exclude the possibility of a line of the expected intensity overlapping the $L$ 911.7 line, although no definite statement can be made on this basis. However, the photo peak of a gamma ray of about 980 keV is seen in the scintillation spectrum of $Bi^{204}$ and appears also in several cases in the coincidence spectra (see e.g. Figs. 7 and 8). Because absorbers of Pb and/or Cd were used in the coincidence experiments, the photo peak at 980 keV should not be due to a sum-up effect of pulses from X-rays and the strong gamma rays of about 900 keV. The transition of 983.6 keV is therefore accepted.

The transitions of 289.3, 374.7, 622.2, 899.2 and 911.7 keV are well known from the decay of 67.5 min $Pb^{204m}$ [6].

## 4. Search for positrons

A search for positrons was undertaken in an intermediate-image beta-spectrometer [15] and the double-focusing spectrometer [13]. The procedure and analysis of the spectra obtained with samples mixed with $Bi^{203}$ and/or $Bi^{205} + Bi^{206}$ are described in our work on $Bi^{203}$ [11]. From the measurements an upper limit for the positron emission in the decay of $Bi^{204}$ could be set to be $5 \times 10^{-4}$ positrons per 374.7 keV transition.

## 5. Scintillation spectrum

Scintillation spectra were measured with a single-channel pulse-height analyzer with automatic recording and a constant relative channel-width of 2 % [14]. The resolution with the $1\frac{3}{4}'' \times 2''$ NaI(Tl) crystal was 9 % for the 662 keV gamma ray

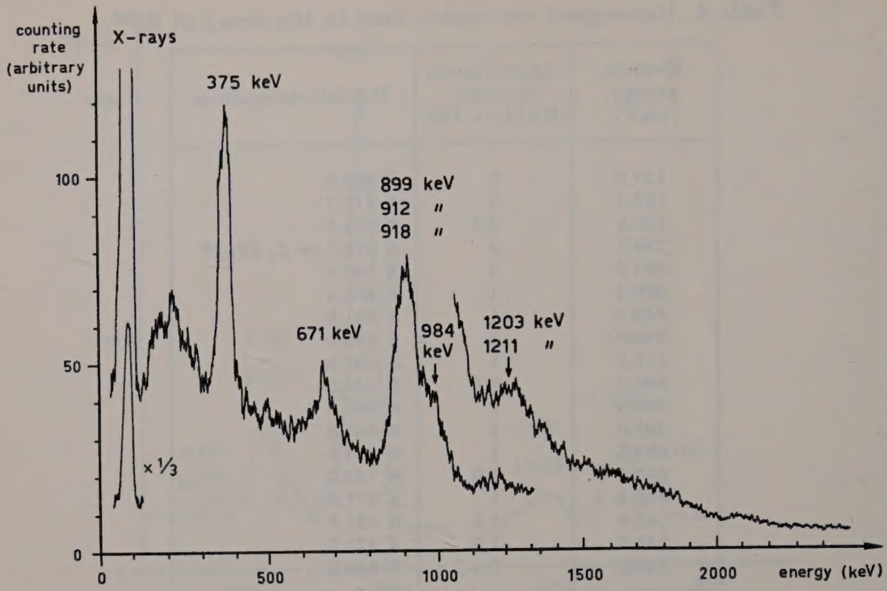

Fig. 4. Scintillation spectrum obtained from two different measurements (low- and high-energy regions) with an electromagnetically isotope-separated sample of Bi$^{204}$. A single-channel pulse-height analyzer with automatic recording and constant relative channel-width of 2 % was used.

of Cs$^{137}$. The spectrum shown in Fig. 4 was obtained from an electromagnetically isotope-separated sample of Bi$^{204}$ [19]. From conversion measurements the contribution from Bi$^{203}$ was estimated to be less than 2 % (cf. Fig. 6 below, and Figs. 10 and 14 in ref. 19). The relative gamma-ray intensities (mean values from several runs) given in Table 2, column 3, were calculated from the photo peaks of the scintillation spectrum and the known energy-efficiency relation for the crystal, and should be considered as rough estimates.

The weak photo peak at about 2 MeV may originate from a gamma ray of this energy, but may also arise from a sum-up of pulses due to the strong 374.7, 899.2 and 911.7 keV gamma rays. As will be found from coincidence measurements, however, the later possibility seems less probable (see Subsection 7.b). The intensity of a gamma ray in the high-energy region would be less than 100 units (cf. Table 2).

An investigation was also made with a hollow crystal (1¾″ × 2″ Harshaw NaI(Tl)). No stronger sum-up peaks were found other than those expected from the well-known transitions of 374.7, 899.2 and 911.7 keV in the isomeric decay.

## 6. Multipolarity assignments and transition intensities

Table 2 includes the multipolarities suggested for some of the transitions, as well as the total intensities of these transitions. The values of the transition intensity $I_{\text{tot}} \approx I_K[1 + 1.3/(K/L) + 1/\alpha_K]$ were calculated by use of the relative $K$-conversion intensities $I_K$, included in Table 1, the theoretical values of $K/L$ given by Rose [20] and the theoretical values of the conversion coefficients $\alpha_K$ given by Sliv and Band [21].

In some cases multipolarity assignments could be suggested from the available data of $K/L$ ratios and conversion in the $L$ sub-shells (see Table 1) by comparison with the theoretical values of Rose [20], and from half-life consideration. In some other cases, the multipolarity assignments could be suggested after use of the additional information given by the relative gamma intensities, included in Table 2. One of these cases is the 670.7 keV transition. Its value of $K/L = 5.8 \pm 0.6$ suggests that the multipolarity is either $E1$ or $M1$, because the theoretical values [20] for these multipolarities are 6.4 and 6.0, respectively. However, the expected gamma intensities for $E1$ and $M1$ are, as can be seen from Table 2, column 2, 4300 and 460, respectively. Thus the measured upper limit of about 800 for the gamma intensity is consistent only with an $M1$ assignment for the 670.7 keV transition.

## 7. Coincidence measurements

Because of the large number of transitions found in the decay of $Bi^{204}$ the main features of the level-scheme could not be unravelled without coincidence experiments.

### (*a*) *Electron-gamma coincidence measurements*

An apparatus for electron-gamma coincidence measurements was set up by use of an intermediate-image beta-spectrometer [15] equipped with an anthracene crystal for the electron detection and a NaI(Tl) crystal for the gamma detection. The resulting pulses were fed to a coincidence apparatus [17] and a 100-channel pulse-height analyzer of the Hutchinson–Scarrott type [16].

To increase the energy resolution of the gamma detector, the crystal was mounted directly on the multiplier. The multiplier system was then placed inside the pole-piece of the spectrometer, and, in order to reduce the influence of the magnetic field on the resolution, a compensating coil was wound around the cover of the multiplier. This arrangement gave nearly the same energy resolution inside and outside the spectrometer (9.5 and 9.0 % respectively for the 662 keV gamma ray of $Cs^{137}$). It can be mentioned that the arrangement with a light-guide and the multiplier outside the spectrometer gave an energy-resolution of 18 %.

The optimum resolving time for the coincidence arrangement was $2\tau = 9$ m$\mu$sec. However, in order to avoid time-wasting trimmings between the different runs, a resolving time of $2\tau = 30$ m$\mu$sec was used. The coincidence apparatus is described in detail by Johansson elsewhere [17].

The procedure of measurements was to focuse a conversion line or line group in the beta channel and collect the whole coincidence spectrum in the 100-channel analyzer. The energy resolution of the beta spectrometer was set to be about 3 %. Singles spectrum was in every case taken before and after the coincidence measurement. It was sometimes checked also during the measurement that no shift had taken place due to instability of the high tension etc.

Between the gamma detector and the source it was possible to introduce suitable absorbers and thus reduce the counting-rate in the gamma channel, which by the electronics (100-channel analyzer) was limited to about 1000 c/sec. This was important especially in experiments with weak lines. X-rays were shielded off by use of 1 mm Cd-plates, and Pb-absorbers of several mm thickness were used when coincidences with gamma rays of higher energies were studied.

From the small resolving time $2\tau = 30$ m$\mu$sec, the chance-coincidence rate is expected to be negligible in the experiments. This was also confirmed by introducing

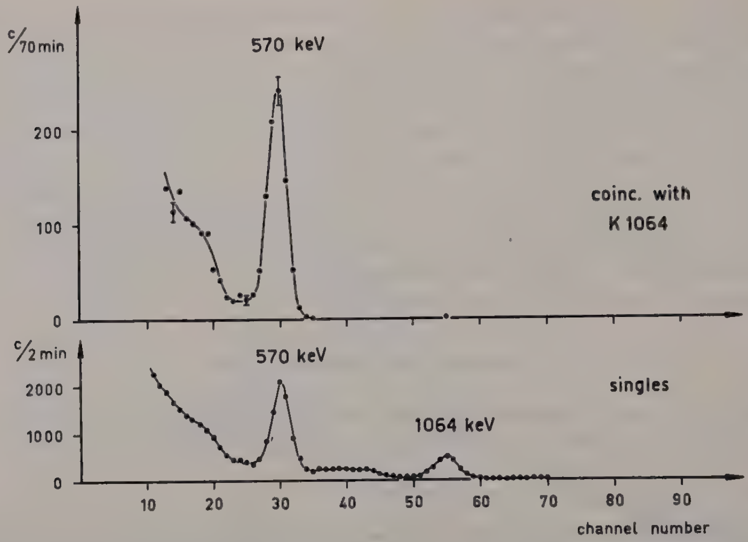

Fig. 5. Spectrum in coincidence with the $K$-conversion electron emission of the 1063.9 keV transition in Bi$^{207}$, and singles spectrum, measured in order to check the apparatus used in the coincidence experiments with Bi$^{204}$. The instruments include a 100-channel pulse-height analyzer and a coincidence apparatus with a resolving time $2\tau = 30$ mμsec. The total chance-coincidence rate (from all channels together) is of the order of 1 c/h.

a large time-delay in one of the channels. A separate experiment was also made with a sample of Bi$^{203}$ feeding the electron channel and a sample of Bi$^{204}$ feeding the gamma channel. The total chance-coincidence rate (sum from all channels) was found to be of the order of 1 c/hour with a counting rate of 800 c/sec in the gamma channel and 300 c/min in the electron channel. This should be compared with a total coincidence rate of the order of 1000 c/hour in the measurements of coincidences in Bi$^{204}$ with about the same counting rates in the two channels as above.

In order to check the coincidence apparatus, a sample of Bi$^{207}$ was used. This isotope exhibits a well-known decay in which the gamma rays of 569.7 and 1063.9 keV, emitted in cascade, are over-all dominating [1]. Measurements were carried out several times with the $K$-conversion line of the 1063.9 keV transition focused in the electron channel. As can be seen from the coincidence spectrum in Fig. 5, the photo peak of the 569.7 keV gamma ray is very pronounced. In the corresponding coincidence spectrum, obtained with the $K$ line of the 569.7 keV transition focused in the beta channel, the photo peak of the 1063.9 keV gamma ray is quite dominating (see also ref. 17).

The sample used in the coincidence measurements on Bi$^{204}$ was obtained from an electromagnetic isotope separation [19]. The amount of Bi$^{203}$ present was estimated to be less than 2 % and had no influence in the measurements (cf. Figs. 10 and 14 in ref. 19, and Fig. 6 below). Fig. 6 shows the conversion line spectrum measured at 3 % resolution with the beta spectrometer used in the coincidence experiments. Because of the complexity of the spectrum coincidences with only the strongest lines could be studied (see Fig. 6 and Table 4).

Fig. 7 shows the coincidence spectrum obtained with the conversion line group

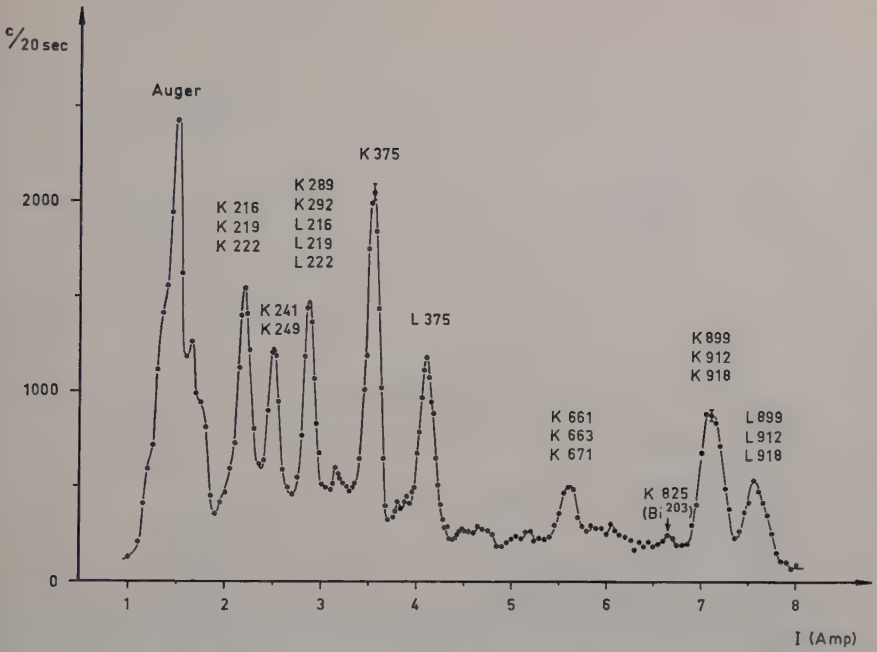

Fig. 6. Conversion line spectrum of $Bi^{204}$ measured at 3 % resolution in an intermediate-image beta-spectrometer used for electron-gamma coincidence experiments. The sample was obtained from an electromagnetic isotope separation. The presence of a very small amount of $Bi^{203}$ ($< 2$ %), revealed by the appearance of $K$ 825.2 (the strongest conversion line in $Bi^{203}$), had no disturbing influence in the coincidence measurements.

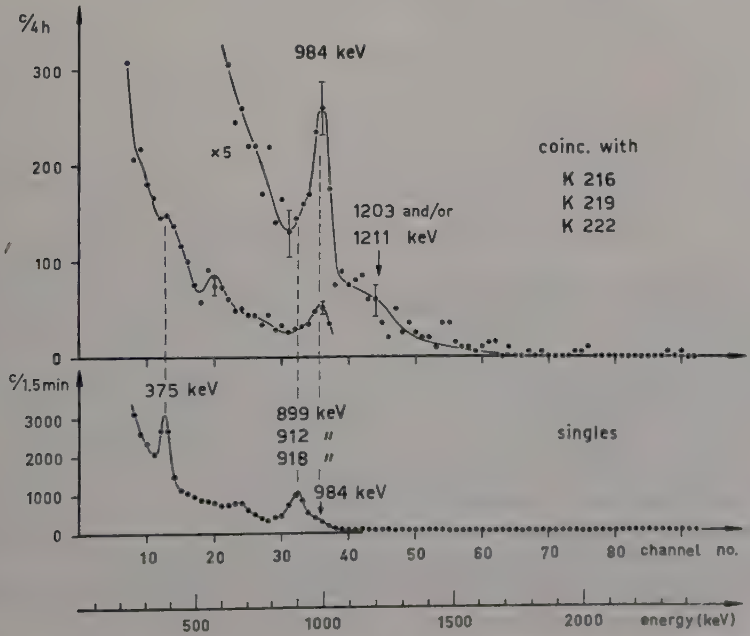

Fig. 7. Singles spectrum of $Bi^{204}$, and coincidence spectrum obtained when the beta spectrometer was set to focuse the conversion line group ($K$ 216.0 + $K$ 219.1 + $K$ 221.9). (cf. Fig. 6). The chance-coincidence rate is negligible (total rate ≈ 1 c/h). A 100-channel pulse-height analyzer was used, and absorbers of 1 mm Cd and 2.5 mm Pb were introduced between the source and the gamma detector. (A special measurement was made for the low-energy region).

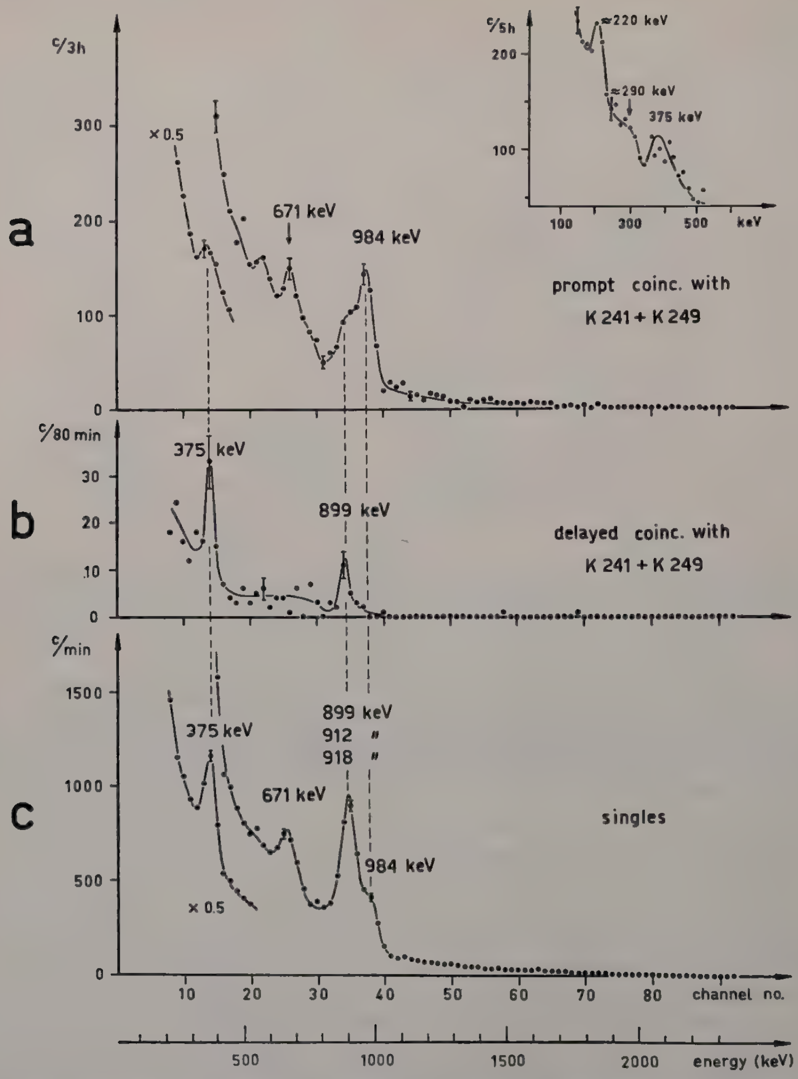

Fig. 8. Coincidence spectra obtained by use of a 100-channel pulse-height analyzer and with the line group ($K$ 240.5 + $K$ 249.0) of $Bi^{204}$ focused in the beta channel (cf. Fig. 6). The chance-coincidence rate (total rate ≈1 c/h) is negligible. 1 mm Cd- and 2.5 mm Pb-absorbers were inserted between the source and the gamma detector. (*a*) Coincidence spectrum measured with no time-delay between the beta and gamma channels. The inset shows part of a special run (with 1 mm Cd-absorber) for the low-energy region. (*b*) Coincidence spectrum obtained by introducing a time-delay of about 0.2 μsec in the beta channel. (*c*) Singles spectrum.

($K$ 216.0 + $K$ 219.1 + $K$ 221.9) focused in the beta channel (cf. Fig. 6). Pb- and Cd-absorbers were introduced between the source and the gamma detector. The photo peak at about 980 keV is assumed to originate from the 983.6 keV transition discussed in Section 3. A special run was made for the low-energy region.

The 980 keV photo peak is also observed in the prompt-coincidence spectrum shown

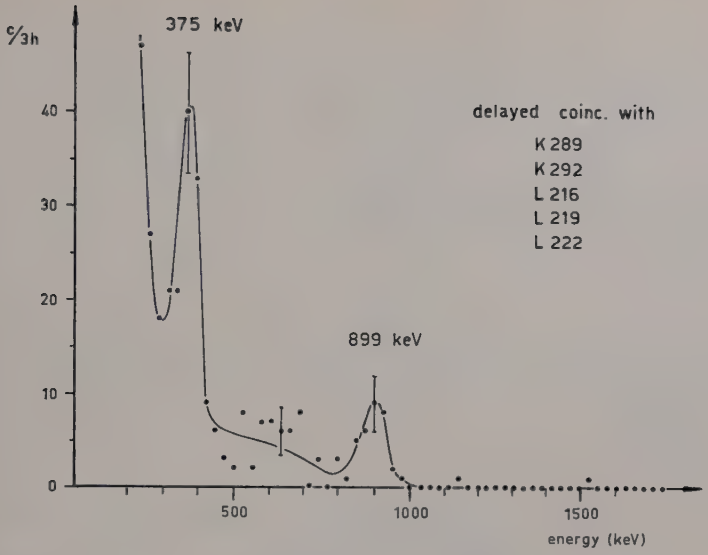

Fig. 9. Delayed-coincidence spectrum obtained with a 100-channel pulse-height analyzer, for the case that the conversion line group, dominated by $K$ 289.1, was focused in the beta spectrometer (cf. Fig. 6). The chance-coincidence rate is negligible (total rate $\approx 1$ c/h). Absorbers of 1 mm Cd and 2.5 mm Pb were inserted between the source and the gamma channel. The time-delay introduced in the beta channel was about 0.2 $\mu$sec. The photo peaks are probably due to the 374.7 and 899.2 keV gamma rays, emitted in cascade from the 0.26 $\mu$sec state in $Pb^{204}$.

in Fig. 8*a*. In this case the spectrometer was set to focuse the line group ($K$ 240.5 + $K$ 249.0), (cf. Fig. 6). Pb- and Cd-absorbers were used also in this case. A special run (with Cd-absorber only) was made for the low-energy region. Part of this spectrum is shown in the inset of Fig. 8*a*.

The delayed-coincidence spectrum, represented in Fig. 8*b*, was obtained by introducing a time-delay of about 0.2 $\mu$sec in the beta channel. As can be seen, only the photo peaks at about 375 and 900 keV appear in this spectrum (observed also in the prompt-coincidence spectrum, although partly overlapped). These photo peaks are obviously due to the gamma rays of 374.7 and 899.2 keV, known to be emitted in cascade from the 0.26 $\mu$sec state in $Pb^{204}$ [6].

A delayed-coincidence spectrum, similar to the one in Fig. 8*b*, is shown in Fig. 9. In this case, the conversion line group dominated by $K$ 289.3 was focused in the beta channel. The spectrum in delayed coincidence with the conversion electron emission mainly from the 670.7 keV transition had also a similar appearance.

In the prompt-coincidence spectra obtained with the $K$ 374.7 line and the ($K$899.2 + $K$ 911.7 + $K$ 917.6) line-group observed in the betaspectrometer, the photo peaks of about 900 and 375 keV, respectively, were very pronounced. Also this result should be due mainly to the cascade of the strong 374.7 and 899.2 keV transitions. In the case when the $K$ line of the 374.7 keV transition was focused in the beta channel, a measurement was also carried out with a time-delay introduced in the gamma channel. Additional photo peaks of about 290, 500, 670, 980 and 1200 keV were then observed.

*Table 4.* Electron-gamma coincidences in the decay of $Bi^{204}$.

| Conversion lines focused in electron channel | Photo peaks visible in the coincidence spectrum (approximate energy values in keV) | |
|---|---|---|
| | Prompt coinc.[a] | Delayed coinc. |
| *K* 216.0 +<br>*K* 219.1 +<br>*K* 221.9 | 250?, 290?, 550, 980, 1200<br>(375, 900) | |
| *K* 240.5 +<br>*K* 249.0 | 220, 290, 550, 670, 980, 1200?<br>(375, 900) | 375, 900 |
| *K* 289.3 +<br>*K* 292.2 +<br>*L* 216.0 +<br>*L* 219.1 +<br>*L* 221.9 | 220, 250, 500, 600–700<br>(375, 900) | 375, 900 |
| *K* 374.7 | 900<br>(290, 500, 670, 980?, 1200) | 290, 500, 670, 980, 1200<br>(900) |
| *K* 660.8 +<br>*K* 663.2 +<br>*K* 670.7 | | 375, 900 |
| *K* 899.2 +<br>*K* 911.7 +<br>*K* 917.6 | 375<br>(290, 670?, 900, 980) | |

[a] The transitions in parentheses in column 2 probably take place on the "opposite side" of the isomeric 0.26 μsec level, as compared with the transitions given in column 1.

A summary of the electron-gamma coincidence information is given in Table 4. Most of the results reported there were checked by repeated measurements. Because of the complexity of the spectra, only qualitative results are given.

### (*b*) *Gamma-gamma coincidence measurements*

After having the beta channel in the apparatus, described in *a* above, replaced by a gamma channel, gamma-gamma coincidence measurements were performed. The discriminator was set to accept pulses due to gamma rays of energy larger than 2 MeV (cf. scintillation spectrum, Section 5 and Fig. 4). The coincidence spectrum shows peaks at about 290, 375, 500, 670 (weak), 900 and 980 (weak) keV. As was discussed in Section 5, the 2 MeV photo peak observed in the scintillation spectrum might be due to a sum-up of pulses due to gamma rays of lower energies. This possibility, however, seems less probable, because the total coincidence rate would in that case have been at least 5 times less than what was actually found (14 c/min).

## 8. Energy levels in $Pb^{204}$

The information available for the construction of the decay scheme of $Bi^{204}$ consists of the accurate energy values of the transitions, the multipolarity assignments and transition intensities of some of them, and the results of the coincidence measurements. Considerable aid should be gained also from the knowledge of the decay scheme of the 67.5 min isomer $Pb^{204m}$ [6]. In addition, some clues might be obtained from theoretical predictions [4, 5] (cf. Fig. 10). However, the large number of gamma rays found in the decay of $Bi^{204}$ leads to great difficulties. Firstly, when starting the construction of the decay scheme by considering the gamma energies only, it is found that, in spite of the energy accuracy, a great variety of combinations of the transitions are possible. Secondly, it is often impossible to make unambiguous interpretations of the coincidence results. Finally, the theoretically calculated level energies should be considered as very rough because of the large approximations introduced in the treatment of a case with as much as four neutron holes.

### (*a*) *Energies of levels*

Fig. 10 shows in addition to the levels of 0, 899.2, 1273.9, 1563.2 and 2185.6 keV, well-known from the isomeric decay [6], six new levels in $Pb^{204}$. These should be considered as tentative.

The energy level of 2476.9 keV is suggested by the following energy relations (in keV):

$$\begin{aligned} 660.8 + 542.3 &= 1203.1 \\ 532.3 + 670.7 &= 1203.0 \\ 412.2 + 791.1 &= 1203.3 \\ 219.1 + 983.6 &= 1202.7 \\ 221.9 + 692.0 + 289.3 &= 1203.2 \\ \text{Energy of cross-over gamma ray} &= 1202.9 \\ \textit{Mean value} &= 1203.0 \end{aligned}$$

No energy sum differs from the mean value of 1203.0 keV by more than 3:10000, but in spite of this it is not excluded that one or several of the sum coincidences are accidental. However, the adoption of the 2476.9 keV level and the inclusion of the above-mentioned gamma rays in the decay scheme, is supported by the fact that a strong feeding to the 1273.9 keV level is required, as can be seen from a consideration of transition intensities. In the decay of the 67.5 min isomer $Pb^{204m}$ the 911.7, 374.7 and 899.2 keV transitions have about the same transition intensities. As can be seen from Table 2 and Fig. 10, however, the relative transition intensities found in the decay of $Bi^{204}$ are quite different. This proves that, in this decay, additional feeding takes place—by transitions which do not originate from the 2185.6 keV 9 − state—to the three lowest excited states, and predominantly to the state of 1273.9 keV.

The inclusion of the gamma rays given in the energy relations above, suggests five new levels between the 1563.2 and the 2476.9 keV levels. The proposed order of emission of the gamma rays in the different cascades seems to be the most probable because of the transition intensities and the possibility of including more gamma rays in the level scheme (see Fig. 10).

According to the coincidence results given in Table 4 at least one of the 216.0, 219.1 and 221.9 keV transitions is in coincidence with a transition of about 1200 keV. If the 219.1 and 221.9 keV transitions are assumed to take place from the 2476.9 keV level, as suggested above, the 216.0 keV transition should be responsible for the coincidence result. Since this transition is not observed in the decay of $Pb^{204m}$, it may take place from a high-lying level and directly or indirectly feed the 2476.9 keV level.

The result obtained when the conversion line group ($K$ 240.5 + $K$ 249.0) was focused in the beta channel, may be understood if also the 240.5 keV transition has

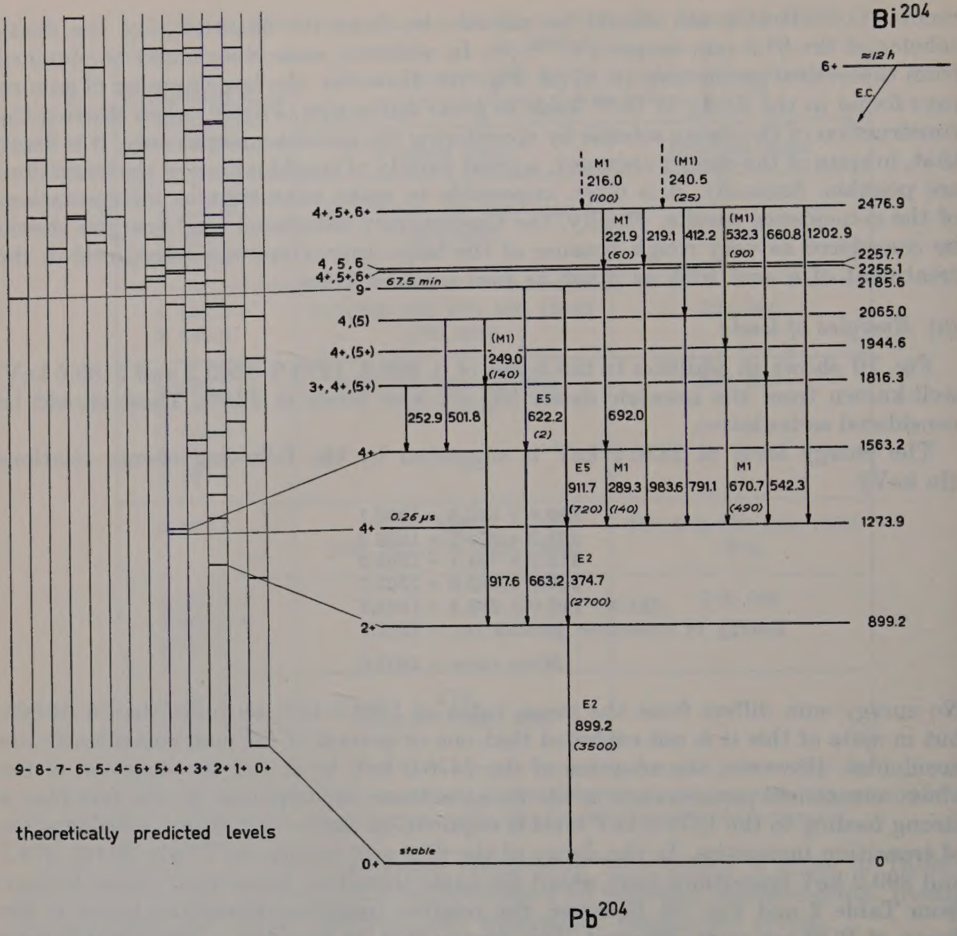

Fig. 10. Tentative level scheme of Pb$^{204}$ as obtained from the decay of Bi$^{204}$. The transitions of 216.0 and 240.5 keV are by coincidence experiments suggested to feed—directly or indirectly—the 2476.9 keV level. In cases where multipolarity information is available the total relative transition intensity has been given below the energy value (cf. Table 2). The position of the Bi$^{204}$ ground state is arbitrarily chosen.

Theoretically predicted levels are shown to the left in the figure. Blomqvist [5] has recalculated True's energy values [4] by use of Pryce's new interaction parameters [24] and has also added some new levels.

such a position that it directly or indirectly feeds the 2476.9 keV level. The possibility that the observed gamma ray of 240.5 keV should be emitted from the 2185.6 keV 9 – state, as may be suggested by the energy relation (in keV) $240.5 + 670.7 = 911.2$ (cf. energy of cross-over gamma-ray = 911.7 keV), must be rejected for intensity reasons. This transition was not detected at all in the decay of Pb$^{204m}$ (i.e. $K\ 240.5 <$ ca 1 % of $K$ 911.7), whereas in the decay of Bi$^{204}$ it appeared with a relatively strong intensity ($K\ 240.5 = 25\,\%$ of $K$ 911.7). For the same reasons the transition of 368.0 keV found in the decay of Bi$^{204}$ cannot be identical with a transition from the 9 – state to the state of 1816.3 keV.

In the study of $Pb^{204m}$ the 1563.2 keV state was found to be de-excited by a 289.3 keV transition. As is indicated in Fig. 10, it might be de-excited also by a 663.2 keV transition. The additional feeding which is proved to take place to the 1563.2 keV level in the decay of $Bi^{204}$ (see above), may explain why the 663.2 keV transition is observed in this decay although not in the isomeric decay (cf. $K$ 663.2 = 5 % of $K$ 289.3).

### (*b*) *Spin and parity assignments*

The spins and parities of the states of 0, 899.2, 1273.9, 1563.2 and 2185.6 keV have been established from studies of $Pb^{204m}$ [6]. The additional spin and parity assignments shown in Fig. 10 seem to be the most probable according to the different sets of data, but should be considered as tentative.

As an example of the difficulties in making definite assignments, one case, the 1944.6 keV state, will be treated here in detail. From the multipolarity $M1$ of the 670.7 keV transition the 1944.6 keV state is suggested to have spin and parity $3+$, $4+$ or $5+$. The $3+$ assignment may, however, be excluded, since no transition is observed from this state to the 899.2 keV $2+$ state. In order to decide which of the remaining possibilities, $4+$ or $5+$, should be chosen, the fact may be considered that no transition of 241.0 keV was observed in the decay of $Pb^{204m}$ from the $9-$ state to a state of 1944.6 keV. Such a transition would be of multipolarity $E5$ or $M4$ for the spin and parity assignment $4+$ or $5+$, respectively. According to an estimate made by use of Sliv's $K$-conversion coefficients [21], Rose's $K/L$ ratios [20] and Weisskopf's theory [22] for transitions of a single proton ($S = 1, r = 1.20 \times 10^{-13} A^{1/3}$), the conversion lines of a 241.0 keV $E5$ transition would all be less than 0.1 % of $K$ 911.7 (cf. $K$ 622.2 = 0.7 % of $K$ 911.7) and would thus be undetected in the measurements. On the other hand, if the multipolarity is assumed to be $M4$, the $K$- and $L$-conversion lines would both be about 40 times stronger than $K$ 911.7, which means that the transition must be at least $10^3$ times slower than expected from this theory to be undetected in the experiments. Although the transitions here may not be due to a single particle, one may expect that the estimates above give the right order of magnitude, since this has generally been found to be the case in this $Z$-region, especially for high multipolarities. For this reason the assignment $4+$ seems preferable, but a $5+$ assignment cannot be excluded. However, the large intensity of the 670.7 keV transition observed in the Bi decay is best understood if direct feeding takes place to the 1944.6 keV state from the ground state of $Bi^{204}$. Since this state has spin and parity $6+$ (spin value 6 recently established by Lindgren [23], using atomic beam resonance techniques), it seems probable that electron capture feeding takes place predominantly to states with spin values 5, 6 or 7. From this point of view the $5+$ assignment seems more probable than $4+$. Thus it is difficult to decide between these two assignments.

It is satisfactory to find that the level scheme of Fig. 10 and the level scheme found independently by Fritsch [10] are in very good agreement. It must, however, be pointed out that both level schemes should be considered as tentative. Because of the complexity of the decay more experiments are needed. The actual case emphasizes the importance of having apparatus with high transmission as well as high resolution and especially electron-electron coincidence apparatus of this kind.

In addition to the experimentally deduced levels Fig. 10 shows some theoretically predicted levels. The energy values given by True [4] have been recalculated by Blomqvist [5] using Pryce's new interaction parameters [24]. Some new energy values calculated by Blomqvist are also given.

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